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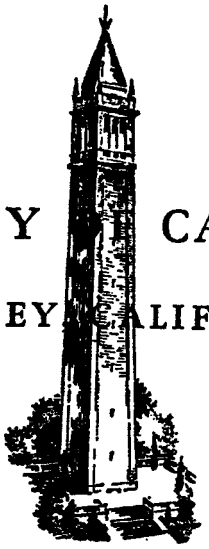

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MEASUREMENTS OF MASS AND MOMENTUM FLUX IN NON-IDEAL MOLECULAR BEAMS

by

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ABSTRACT

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Molecular beams formed by effusion through an orifice whose size is of the same order as the mean free path in the source chamber are non-ideal in the sense that the effects of intermolecular collisions in the beam are not negligible.

The properties of such beams have been investigated by measuring, over a wide range of source conditions, the normal momentum transferred from the beam to a diffusely reflecting test surface mounted on a sensitive torsion balance. The perturbation of the measured force on the test surface from its ideal free molecule value is interpreted in terms of the mass and momentum flux perturbations in the beam and compared with the predictions of recent theories. A substantial discrepancy is noted between the calculated and measured perturbations.

These experiments have demonstrated the feasibility of making absolute measurements of momentum transfer in particle-surface interactions.

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1.0 INTRODUCTION

A previous report from this laboratory[1] has described the development and use of a molecular beam and torsion balance apparatus to study normal momentum transfer in particle-surface interactions. The main objective of this work was to measure the force exerted on a test surface by an incident beam of gas molecules as a function of the surface temperature, and hence to obtain information concerning the manner in which the gas molecules were re-emitted from the surface. The work was largely successful and many interesting results were obtained. It was shown that the measured values of momentum transfer to a heated surface could be explained on the basis of a model in which the reflection of molecules at the test surface was assumed to be perfectly diffuse, although the energy accommodation was not always complete. The accommodation coefficients measured varied between about 0.5 for light gases such as helium, and close to unity for argon and carbon dioxide, and were relatively insensitive to the material of the test surface, and the surface temperature.

There were, however, a few minor deficiencies in the work which remained unresolved. Perhaps the most important of these was the fact that no absolute measurements of momentum transfer were made; the conclusions above were all based on relative measurements between a heated and an unheated surface. A crucial assumption in the interpretation of the results was that in the case of the unheated surface (whose temperature was therefore equal to the gas source temperature) the normal momentum carried away from the surface by the re-emitted molecules was equal to two thirds of the incident momentum. This would be true in the case of a diffusely reflecting surface and an ideal Maxwellian incident beam. However, the source pressures at which most of the experiments were performed were much too high for molecular collisions in the source chamber to be neglected,

so that in all probability the incident beam was markedly non-Maxwellian. Furthermore, no direct experimental evidence was obtained to confirm molecular reflection at the unheated test surface was in fact diffuse (although the results of other workers suggest that this is true for most polycrystalline materials).[2]

The work described in this report was aimed at modifying the apparatus in such a way as to make feasible absolute measurements of momentum flux in the incident beam and of momentum transfer to a test surface, thereby removing the main shortcoming of the earlier work. It was also hoped that in the course of new experiments light would be shed on the question of diffuse reflection at the test surface under the present experimental conditions so that the validity of Stickney's assumptions might be clarified. It was hoped to examine this matter by two procedures:

1. By making measurements of momentum transfer at sufficiently low source pressures to allow the properties of the incident beam to be calculated on the basis of elementary kinetic theory (free molecule limit).

2. By comparing the momentum transfer to a test surface over a wide range of source pressures, with that to a "momentum trap" specifically designed to give completely diffuse reflection.

Moreover, since the problem of orifice flow is an important aerodynamic problem in its own right, it was hoped to use the torsion balance to investigate the manner in which the properties of the molecular beam were dependent on the magnitude of the source pressure. This would also have the practical value of allowing the effect of the non-ideal beam in the earlier work to be estimated.

2.0 SUMMARY OF PREVIOUS WORK

2.1 Apparatus

The molecular beam and torsion balance apparatus is described in detail in Ref 1, and only a very brief description of the main features of the apparatus will be given here Figure 1 shows a schematic diagram of the apparatus and is largely self-explanatory. The design differs from that of a conventional molecular beam system in only two ways.

1. The defining and test regions share the same pumps. This was made possible by the use of a high speed 6-inch diffusion pump backed by a 5 HP Kinney mechanical pump, and by the nature of the problem.

2. In the work described in Ref. 1 the beam was not well collimated, but was allowed to cover one-half of the test surface. The advantage of this system is that it gives the maximum possible rate of momentum transfer to the surface. However, as will be shown later, it also suffers from disadvantages which led to its being abandoned in the present work

Figure 2 shows a diagram of a torsion balance. This consisted of a 2 cm × 1 cm sheet of metal foil (usually about 0.001 inch thick, which was mounted symmetrically on a 0.006 inch diameter quartz rod which also carried a small galvanometer mirror whose plane coincided with that of the surface. This was used in conjunction with a lamp and scale to sense the angular position of the test surface. The balance was suspended from a torsion head by means of a 0.0002 inch diameter tungsten torsion fiber. The whole balance was grounded to prevent the build-up of electrostatic charge which would cause erratic behavior.

The temperature of the test surface could be varied up to about 600°C by means of radiation heating from a 500 watt projector.

2.2 Procedure

Suppose the total momentum flux incident upon the heated test

surface is p_i and that carried away is p_r. The total force acting on the surface is $p_i + p_r$, and this will be proportional to the angular deflection, θ, of the torsion head which is required to bring the test surface back to its zero position. Hence

$$p_i + p_r = k\theta \tag{2.1}$$

In the special case where the surface is unheated, denoted by the subscript o

$$p_{i_o} + p_{r_o} = k\theta_o \qquad \text{or} \qquad 1 + \frac{p_{r_o}}{p_{i_o}} = \frac{k\theta_o}{p_{i_o}} \tag{2.2}$$

If we assume that in the latter case all molecules are reflected diffusely at the surface it follows that for an ideal Maxwellian incident beam $p_{r_o} = \frac{2}{3} p_{i_o}$ irrespective of the degree of energy accommodation, since the source and test surface temperatures are equal. From (2.2) we therefore obtain

$$\frac{5}{3} = \frac{k\theta_o}{p_{i_o}} \tag{2.3}$$

Dividing Eq. (2.1) by Eq. (2.3)

$$1 + \frac{p_r}{p_i} = \frac{5}{3} \frac{\theta}{\theta_o} \frac{p_{i_o}}{p_i} \tag{2.4}$$

Now since the beam is assumed to be ideal the incident momentum flux p_i will be proportional to the source pressure P (see Eq. 4.1) so that Eq. (2.4) becomes after rearrangement

$$\frac{p_r}{p_i} = \frac{5}{3} \frac{\theta}{\theta_o} \cdot \frac{p_o}{P} - 1 \quad . \tag{2.5}$$

Hence the ratio of p_r to p_i may be deduced either from measurements of the deflection θ keeping the source pressure constant or measurements of

source pressure P keeping the deflection constant. Each method has its advantages and shortcomings which are discussed in Paragraph 3.1(c). Both methods were used in the previous work and gave good agreement.

2.3 Results

The measurements of p_r/p_i described in Ref. 1 were satisfactorily explained in terms of the following model in which reflection of molecules at the test surface is assumed to be completely diffuse. Consider the two extreme cases of complete and zero energy accommodation. In the first case it may be shown that

$$p_r = \frac{2}{3} p_i \left(\frac{T_w}{T_i}\right)^{1/2} \tag{2.6}$$

for an ideal beam. In the second case

$$p_r = \frac{2}{3} p_i \tag{2.7}$$

In the general case the reflected momentum will be somewhere between these two values, so we may define a normal momentum transfer coefficient σ'' such that

$$\frac{p_r}{p_i} = \frac{2}{3}\left[(1 - \sigma'') + \sigma'' \left(\frac{T_w}{T_i}\right)^{1/2}\right] \tag{2.8}$$

Clearly $\sigma'' = 1$ for complete energy accommodation and $\sigma'' = 0$ for zero energy accommodation.

Combining Eq. (2.8) with Eq. (2.5) and calling $\theta/P = F$ so that the following is applicable to both the constant pressure and constant torque procedures we have

$$\frac{5}{3}\left(\frac{F}{F_o} - 1\right) = \frac{2}{3}\left[(1 - \sigma'') + \sigma'' \left(\frac{T_w}{T_i}\right)^{1/2}\right] \tag{2.9}$$

or

$$\frac{F}{F_o} - 1 = \frac{2}{5}\,\sigma'' \left[\left(\frac{T_w}{T_i} \right)^{1/2} - 1 \right] \tag{2.10}$$

Theoretically it is possible to make an absolute determination of σ'' for any gas-surface combination by simultaneous measurement of F/F_o and T_w/T_i. In practice, however, T_w is extremely difficult to measure since the presence of thermocouple connections to the surface would disturb the operation of the torsion balance and the temperatures used (~ 500°C) are too low to be measured by optical means. In order to circumvent this difficulty Stickney was obliged to estimate T_w indirectly from experimental momentum transfer measurements made with argon which was assumed to be completely accommodated to the surface temperature (possible justifications for this assumption are discussed in Ref. 1, p. 32). His measured values of σ'' are therefore relative values referred to that of argon as unity. These vary between about 0.5 for light gases such as helium and hydrogen and ~ unity for carbon dioxide and nitrogen. The nature of the test surface appeared to have little or no effect on the magnitude of σ'' for a given gas--a fact which suggests that surface contamination played an important role in these measurements. For all gas-surface combinations investigated σ'' appeared to be independent of surface temperature within the experimental error. This gives support to the basic validity of the interaction model outlined above and shows σ'' to be a useful parameter for describing such interactions.

3.0 PRESENT INVESTIGATION. EXPERIMENTAL

3.1 Modifications to Previous Apparatus

The apparatus in the form in which it was used in the previous work was unsuitable for absolute determinations of beam momentum and mass fluxes for a variety of reasons.

a) The apparatus can measure only the sum of the incident and reflected momenta $p_i + p_r$. In order to determine p_i separately the relationship between p_r and p_i must be known (e.g., is the reflection diffuse or specular, and if the former, is the energy accommodation complete or not). In the case of an experimental test surface this relationship is not known and can only be hypothesized.

b) The solid angle subtended at the source by the test surface is difficult to determine accurately as the distance between the test surface (which is freely suspended) and the source is very difficult to measure.

c) It is virtually impossible to construct and mount a balance in such a way that the axes of the torsion head, the torsion fiber and the test surface all coincide. This means that the moment arm of the beam about the torsion axis will vary with the torsion angle and the latter will not be strictly proportional to the momentum transfer rate. On the other hand, the constant torque procedure which is designed to bypass this difficulty suffers from the disadvantage that unless the beam is ideal (which is true only for very low source pressures) the momentum flux is not directly proportional to the source pressure. The fact that in the previous work measurements obtained by the two procedures were generally in good agreement may be taken as indirect proof that these effects are not large. Nevertheless, for absolute measurements it was considered desirable to eliminate them completely.

It was decided to attempt to overcome difficulty a) by mounting a momentum trap on the balance in place of a plane test surface. This trap was designed so that all molecules entering it had a sufficiently large number of collisions with the interior walls to insure thermal equilibration and subsequent Maxwellian re-emission. The design of the trap is discussed in detail later. Meanwhile it is necessary to remark that the entrance to the trap consisted of a vertical slit 1 cm long and 2 mm wide and the solid angle subtended by it at the source ($\sim 10^{-3}$ steradian) was small enough to be considered elemental. It was obviously necessary to make sure that the couple acting on the balance was due entirely to molecules which entered the momentum trap and were completely accommodated. This was accomplished by introducing a defining slit of appropriate and accurately known dimensions between the source and the entrance to the trap. This slit was rigidly mounted to the source chamber so that the slit-source distance could be accurately measured before assembling the apparatus. This arrangement therefore overcame the difficulty described in b).

Difficulty c) was solved by measuring the moment arm at each experimental reading by means of a cathetometer. The exact manner in which this was done will be described later (Sections 3.1.8 and 3.1.9).

3.2 Calibration of the Torsion Balance

One further requirement for absolute measurements is that the torsional constant of the fiber be accurately known. The only feasible method of determining this quantity consists of suspending from the fiber a body of known moment of inertia and measuring the period of the resulting torsional oscillations. This presented difficulties as the momentum trap balance was of irregular shape and consequently its moment of inertia was difficult to calculate. On the other hand, the possibility of measuring

the torsional constant of a fiber before attachment to the balance was considered unreliable. The following method was eventually adopted. The balance was constructed in its final form with torsion fiber in place and a fine copper needle was attached in a horizontal position to the back of the mirror mounting plate by means of silver paint. The moment of inertia of this needle about a perpendicular axis through its center was of the same order as that of the balance. The mounting operation was performed under a microscope to insure that the center of the needle coincided accurately with the point of attachment of the torsion fiber. The balance was then suspended in the apparatus and its periodic time was measured (this involved evacuation of the test chamber to eliminate the effect of air damping). Following this the balance was removed from the apparatus and the copper needle carefully detached. The balance was then re-installed in the apparatus where, after its new period of oscillation had been determined, it remained for the rest of the measurements. Finally, the copper needle was carefully weighed, its length measured on a travelling microscope and hence its moment of inertia calculated. Sufficient information was then available to allow the moment of inertia of the balance and the torsional constant of the fiber to be calculated.

3.3 Design of the Momentum Trap

Diagrams indicating the method of construction of the momentum trap are shown in Fig. 3. The material used was 0.001 inch aluminum foil, folded as shown, and cemented in position by means of Eccobond solder 58C. In addition, a piece of crinkled foil was inserted along the back wall of the trap chamber to increase the diffuseness of molecular reflection there. The trap was made symmetrical about the quartz fiber axis for reasons of dynamic balance. The small tabs above and below the entrance slit indicated

the position of the slit when the trap was viewed from behind, and were used for alignment purposes. A diagram of the complete balance is shown in Fig. 4. Its moment of inertia was determined by the method described in Section 3.2, and was found to be

$$6.25 \pm 0.03 \times 10^{-2} \text{ gm cm}^2 \quad .$$

3.4 The Defining Slit

The defining slit was made from pieces of 0.005 inch shim stock cemented onto a circular brass plate which had a wide slit milled across the center. The pieces of stock which formed the two long edges of the slit were made by dividing a single sheet of stock and matching the new edges formed. Thus although the slit was not perfectly straight, its width varied less than 0.004 inch (one part in 60). The mean width was determined by measurement with a travelling microscope to be $5.93 \pm 0.02 \times 10^{-2}$ cm and its length 5.074×10^{-1} cm with negligible error. The slit was screwed down over a 1 inch diameter hole in a brass mounting plate which replaced the stainless steel disc used to define the beam in the earlier work. This plate could be rotated from outside the system about two different pivot points, thus allowing the slit to be traversed while remaining vertical. The same partition wall was used.

3.5 The Source Aperture

Most of the previous measurements were made using slit source. However, it was decided that for the present work a circular orifice would be preferable, since then it was possible to use the defining slit to limit the beam in the vertical direction as well as the horizontal. With an extended vertical slit source some molecules could have passed through the defining slit without entering the momentum trap. The source consisted

of a circular hole drilled in a piece of 0.001 inch shim stock. Its mean diameter determined from four travelling microscope measurements was

$$1.010 \pm 0.005 \times 10^{-1} \text{ cm} \quad .$$

In the original form of the apparatus the torsion axis and source axis intersected so that in order to obtain any moment about the torsion fiber the first measurements were made using a beam which was about 10° off the source axis, giving a moment arm of 1 cm. Later, however, the apparatus was modified to allow a limited rotation of the source and defining slit assembly about a vertical axis. This made it possible to use axial beams and hence made comparison between theory and experiment more direct (see Section 5.4).

3.6 The Cathetometer

The cathetometer was manufactured by the Gaertner Scientific Corporation (serial number 1020A). The telescope could be traversed in both horizontal and vertical directions and both coordinates determined to a least count of 0.001 inch by means of Vernier scales.

3.7 The McLeod Gauge

A McLeod gauge was used to measure the source chamber pressure. It had a sensitivity of 1.10×10^{-3} torr/cm^2 and a least count of 2×10^{-2} cm.

3.8 Alignment of the Molecular Beam

The various alignment configurations used are depicted schematically in Fig. 5. Accurate alignment of the apparatus was made possible by the fact that a small glass window was set in the back wall of the source chamber directly opposite the source aperture. By placing a light source

behind this window just outside the test chamber it was possible to view the source aperture through the defining slit.

For the case of the platinum test surface (Figs. 5a and b), the alignment procedure was fairly straightforward since the actual point of impact of the beam on the surface was not critical. The cathetometer was adjusted until the image of the source coincided with the cathetometer cross hairs and the center of the image of the defining slit. In this fashion the beam axis was defined. The galvanometer lamp was then adjusted so that the light spot appeared at the zero point on the scale when the surface was perpendicular to this axis. This procedure was the same for both off-axis and axial beam measurements.

The momentum trap, Fig. 5c, was more difficult as it was necessary to insure that the entrance slit of the trap lay on the beam axis when the balance was perpendicular to it. The following procedure was devised. The defining slit was first swung away to one side to give an unobstructed view of the source, and the balance was raised above the normal operating position so that when viewed through the cathetometer the tab below the entrance slit appeared slightly higher than the source. The balance was then started oscillating about the zero point (which could be determined approximately) and the cathetometer adjusted until during the course of a swing the tab appeared to move out to a position directly above the source and then move back in again. The cathetometer and source then lay along the required beam axis and the defining slit was swung back and adjusted into alignment. Finally, the balance was lowered into the correct operating position and the galvanometer lamp adjusted as before. No axial measurements were made with the momentum trap since the source assembly could not be rotated from outside the test chamber and hence accurate alignment of the momentum trap on the source axis was impossible.

3.9 Experimental Procedure

As stated previously, the constant pressure technique was used throughout the present work. The experimental procedure was as follows:

1. The balance was installed, the test chamber evacuated to about 2×10^{-5} torr and the apparatus aligned.
2. The period of oscillation of the balance was checked and the reading of the cathetometer when sighted along the beam axis was noted.
3. The balance was set to the zero position and the reading of the torsion head was noted.
4. Gas was admitted to the source chamber via a metering valve. When a steady source pressure had been achieved the torsion head was manipulated to bring the galvanometer spot once more to the scale zero point. (In fact, the spot was usually not at rest but oscillated about the zero point with amplitude 1 cm or less.)
5. The new torsion head reading and the McLeod gauge reading were taken and noted.
6. The cathetometer was traversed and sighted on the torsion fiber and the new reading of the horizontal scale was noted. The difference between this reading and the beam axis reading was a measure of the moment arm of the beam about the torsion axis. Since the balance was usually swinging slightly with amplitudes of a few thousands of an inch at the bottom of the torsion fiber it was easier to sight the cathetometer at the top of the fiber. This meant, however, that the cathetometer had to be very carefully leveled before the experiment.

7. The temperature was measured by a mercury thermometer immediately outside the test chamber, and was assumed to be uniform throughout the apparatus. (The temperature was needed only for the calculation of the source Knudsen number, and even then its effect was very small.)
8. Steps 4, 5, 6, and 7 were repeated for different source pressures between 10 and 200 μ Hg.
9. The zero reading of the torsion head was again checked.

The results were plotted in dimensionless form with the ratio p_R of the measured force to the calculated free molecular value for diffuse reflection as ordinate and the inverse source Knudsen number a/λ as abscissa.

4.0 THEORETICAL CONSIDERATIONS

In order to interpret measurements of momentum transfer in terms of the mass and momentum fluxes in the incident beam it is necessary to consider the interaction between the beam and the test surface in some detail. In the following analysis it is assumed that the system is isothermal, i.e., that the source and test surface temperatures are equal.

In a well collimated axial molecular beam operating under free molecule ("ideal") conditions the mass flux $\dot{m}_{FM}$ and the momentum flux $\dot{\mu}_{FM}$ are given, according to elementary kinetic theory, by

$$\dot{m}_{FM} = \frac{\rho\, \bar{c}\, d\Omega\, dS}{4\pi} \qquad \dot{\mu}_{FM} = \frac{3\rho\, RT\, d\Omega\, dS}{4\pi} \tag{4.1}$$

in the usual notation. R is the gas constant/unit mass, $d\Omega$ the solid angle of collimation, and dS the area of the source aperture.

Hence

$$\frac{\dot{\mu}_{FM}}{\dot{m}_{FM}} = \frac{3RT}{\bar{c}} = \frac{3}{2}\left(\frac{\pi RT}{2}\right)^{1/2} \tag{4.2}$$

Under non-ideal conditions we may write

$$\dot{\mu}_i = \dot{\mu}_{FM}\,[1 + \varphi_2(K)] \qquad , \qquad \dot{m}_i = \dot{m}_{FM}\,[1 + \varphi_1(K)] \tag{4.3}$$

where φ_2, φ_1 are the (dimensionless) perturbations of the momentum and mass fluxes from their free molecule values and are functions of the source Knudsen number K. It should be emphasized that φ_2 and φ_1 are not necessarily equal. In fact, experiments have shown[7] that the mean thermal speed of the beam increases with source pressure so that $\varphi_2 > \varphi_1$.

If the beam now strikes a perfectly accommodating surface at normal incidence the molecules will be re-emitted diffusely with a Maxwellian velocity distribution. It may easily be shown that the ratio of the normal momentum flux to the mass flux for such a beam is given by

$$\frac{\dot{\mu}_r}{\dot{m}_r} = \frac{2RT}{\bar{c}} = \left(\frac{\pi RT}{2}\right)^{1/2} \tag{4.4}$$

Now in the steady state the number of molecules striking the surface must be equal to the number leaving, so that

$$\dot{m}_i = \dot{m}_r \quad .$$

Hence

$$\begin{aligned} \dot{\mu}_r &= \left(\frac{\pi RT}{2}\right)^{1/2} \dot{m}_r = \left(\frac{\pi RT}{2}\right)^{1/2} \dot{m}_i \\ &= \left(\frac{\pi RT}{2}\right)^{1/2} \dot{m}_{FM}\,[1 + \varphi_1(K)] \qquad (4.5) \\ &= \frac{2}{3}\,\dot{\mu}_{FM}\,[1 + \varphi_1(K)] \end{aligned}$$

The total normal force p exerted on the surface is given by

$$p = \dot{\mu}_i + \dot{\mu}_r = \dot{\mu}_{FM} [1 + \varphi_2(K)] + \frac{2}{3} \dot{\mu}_{FM} [1 + \varphi_1(K)]$$

$$= \frac{5}{3} \dot{\mu}_{FM} \left(1 + \frac{3\varphi_2 + 2\varphi_1}{5}\right) \tag{4.6}$$

Thus the perturbation of the force on such a surface from its free molecule value is a weighted mean of the momentum and mass flux perturbations in the beam.

Consider now a different type of reflection in which the molecules are re-emitted in perfectly diffuse fashion but with no energy accommodation at the surface. More specifically, assume that each molecule striking the surface retains its own speed but is re-emitted in a random direction. Simple calculation shows that in this case the average value of the normal momentum carried away by each molecule is equal to two thirds of its incident momentum. Hence in this case

$$\dot{\mu}_r = \frac{2}{3} \dot{\mu}_i \tag{4.7}$$

and

$$p = \dot{\mu}_i + \dot{\mu}_r = \frac{5}{3} \dot{\mu}_i = \frac{5}{3} \dot{\mu}_{FM} (1 + \varphi_2) \quad .$$

In this case, therefore, the perturbation of p is equal to that of $\dot{\mu}_i$.

If in the case of a real surface the interaction may be described in terms of a model in which a fraction σ'' (defined by Eq. 2.8) of the incident molecules is completely accommodated to the surface while the remainder are reflected in the manner of the second model (sometimes called multi-specular reflection) the total force acting on the surface will be

$$p = \frac{5}{3} \dot{\mu}_{FM} \left[\sigma'' \left(1 + \frac{3\varphi_2 + 2\varphi_1}{5}\right) + (1 - \sigma'')(1 + \varphi_2)\right] \tag{4.8}$$

which may also be written

$$p = \frac{5}{3}\dot{\mu}_{FM}\left[1 + \frac{3\varphi_2 + 2\varphi_1}{5} + \frac{2}{5}(1 - \sigma'')(\varphi_2 - \varphi_1)\right] \tag{4.9}$$

It is convenient to work in terms of a dimensionless force ratio p_R defined as p/p_{FM} where $p_{FM} = \frac{5}{3}\dot{\mu}_{FM}$.

Hence

$$p_R = 1 + \frac{3\varphi_2 + 2\varphi_1}{5} + \frac{2}{5}(1 - \sigma'')(\varphi_2 - \varphi_1) \tag{4.10}$$

The model predicts, therefore, that the force ratio p_R will depend not only on K but also on σ'', even under isothermal conditions. The magnitude of the effect of σ'' depends on the difference between φ_1 and φ_2. It will be shown later, however, that $\varphi_2 - \varphi_1$ is so small that the variation of p_R with σ'' is virtually undetectable in the present experiments.

5.0 EXPERIMENTAL RESULTS

5.1 The Momentum Trap

Experimental results obtained with the momentum trap are shown in Fig. 6. In this case the abscissa is on a logarithmic scale. For low values of $\epsilon = K^{-1}$ for He, i.e., the free molecule limit the experimental points approach the expected value of unity to within 1 per cent. For argon, whose mean free path is approximately one third that of helium, the results do not extend to low enough values of ϵ to allow a similar statement to be made. However, if the points are plotted on a linear scale and the experimental curve extrapolated by eye to $\epsilon = 0$ it again lies within 1 per cent of the value of unity. The helium and argon results agree fairly closely over their common range when plotted in this manner despite their large discrepancy in mean free paths.

5.2 Platinum Surface

In Fig. 7 the results obtained with helium and argon on platinum are added and seen to be in close agreement with the momentum trap results, within the experimental spread which is typically ±1.5 per cent. This must be taken to indicate that both argon and helium are reflected diffusely from a platinum surface under the conditions of the present experiment. There is no evidence that the experimental points for helium on platinum lie any higher than those taken with the momentum trap even though the value of σ'' for this combination was estimated by Stickney to be about 0.5. One must assume, therefore, that the effect of σ'' on the momentum transfer is too small to be detected in these experiments and therefore φ_2 and φ_1 are very nearly equal. Further evidence supporting this hypothesis will be given later.

5.3 Axial Beam Measurements on Platinum

The results obtained with axial beams of helium and argon on platinum are shown in Fig. 8. The only difference from the previous results is that the perturbation from the free molecule limit is slightly larger than for the off-axis beams. The argon and helium results again agree quite well in their common range.

From Fig. 8b it can be seen that for small values of K^{-1} ($\lesssim 0.5$) the behavior of p_R may be represented by an equation of the form

$$p_R = 1 + A/K \qquad (5.1)$$

The value of A has been estimated by drawing in by eye the best fit asymptote to the experimental curve at the origin. The value obtained is

$$A = 0.28 \pm 0.02 \quad .$$

5.4 Comparison Between Theory and Experiment

It is interesting to compare the axial beam measurements with existing theories of near free molecule orifice flow. This problem was first considered theoretically by Narasimha[3] who was, however, chiefly concerned with obtaining an expression for the total mass flow to compare with the experimental results of Liepmann.[4] By performing one iteration on an integral form of the kinetic equation, using the Krook collision model, Narasimha obtained a first order correction to the free molecule distribution function at the center of a circular orifice which was strictly applicable only to molecules travelling along the axis of the orifice. However, by making reasonable assumptions concerning the distribution function of molecules travelling in non-axial directions, and assuming the local mass flux to be constant over the area of the orifice Narasimha obtained an expression for the total mass flux which was in reasonable agreement with Liepmann's measurements. Using Narasimha's distribution function it is easy to calculate the mass and momentum fluxes in a well collimated axial beam, assuming that molecular collisions downstream of the orifice are negligible. The resulting expressions are

$$\dot{m} = \dot{m}_{FM}\left(1 + \frac{A_1}{K}\right) \quad , \qquad \dot{\mu} = \dot{\mu}_{FM}\left(1 + \frac{A_2}{K}\right) \tag{5.2}$$

where $A_1 = 0.601$, $A_2 = 0.674$. This gives an expression for p_R of the same form as Eq. (5.1), but a value of A of 0.645, which is much larger than present measured value.

Morton[5] has performed a similar calculation with the specific objective, however, of determining the velocity distribution function of a well collimated axial beam well downstream of the orifice. He obtains a value of A_1 of 0.825, but unfortunately does not calculate the momentum flux. Comparing Morton's and Narasimha's values of A_1, it appears that

the effect of downstream collisions is to enhance rather than diminish the intensity of the beam.

Very recently Willis and Fitzjarrald (private communication) have performed a more detailed calculation in which they restrict themselves to the evaluation of mass and momentum flux in a well collimated beam far downstream of the orifice but no longer assume the distribution function to be uniform over the orifice area. Their calculated values of the constants A_1 and A_2 are 0.592 and 0.692, respectively, when using Morton's interpretation of mean free path in terms of collision frequency. Willis,[6] however, regards a rather different interpretation as being more appropriate for comparison between theory and experiment, and this gives $A_1 = 0.465$, $A_2 = 0.543$, and therefore $A = 0.512$, about 80 percent higher than our experimental value.

All the above results assume a gas viscosity proportional to temperature and Prandtl number of unity. Willis and Fitzjarrald find, however, that the use of a more realistic viscosity temperature law reduces the value of A by only 2 percent for helium and even less for argon.

The effect of assuming a Prandtl number of unity (rather than the correct value of 2/3 for a monatomic gas) has not yet been estimated, but it is extremely unlikely that it would account for such a large discrepancy between the experimental and theoretical values of A.

The reason for this discrepancy is not known. It could be due either to the use in the theory of a simplified model of the kinetic equation (the Krook model), or to some spurious effect in the experiment. In the latter case, the most obvious possibility is that the increase in test chamber pressure with source pressure could cause attenuation of the beam at low Knudsen numbers (the excellent agreement in the free molecule limit indicates that attenuation is negligible at the lowest source pressures).

It was decided, therefore, to investigate this point thoroughly. The test chamber pressure as measured by a nude ionization gauge mounted just below the chamber was of the order 2.5×10^{-6} torr when the beam was turned off, but rose to 5.5×10^{-5} torr when the source pressure was 150 μ. At this pressure the mean free path of argon is about 90 cm. Since the distance traversed by the beam between the source and test surface is 6 cm, the beam could have been attenuated by about 6.5 percent. Although this is far too small an effect to explain the discrepancy between theory and experiment, it is nevertheless large enough to warrant correction. It must be emphasized, however, that the calculation above is a very approximate one and could not itself be used as a correction. It merely indicates that it is possible that a significant portion of the beam was lost through attentuation. Whether this was actually the case, and to what extent, can be determined only by experiment.

The simplest means of obtaining a correction is to make momentum flux measurements at different pumping speeds and extrapolate the results to infinite pumping speed, i.e., zero test chamber pressure. An approximate calculation indicated that the pumping speed obtained from the diffusion pump was about 400 liter/sec, whereas its rating was about 1,400 liter/sec. This loss of speed was traced, at least in part, to the presence of a liquid nitrogen trap of low conductance immediately above the diffusion pump. The trap was therefore removed from the apparatus and replaced by a straight through pipe of equivalent length, with the effect of increasing the pumping speed to about 800 liter/sec and of decreasing the presumed attenuation of the beam to approximately half its former value.

Measurements taken with this higher pumping speed are shown in Fig. 9, and compared with the previous results. Within the experimental

error there is no detectable difference between the two sets of data, a fact which can only mean that attenuation was not important in either case.

A second possibility, which is much harder to investigate, is that some molecules after striking the defining plate close to the slit were being re-emitted into the path of the oncoming beam and hence causing attenuation. This effect would presumably be independent of pumping speed and hence could not be detected by the above technique. Willis (private communication) has estimated the size of the effect as about 1/3 per cent for the present geometry of the apparatus. This is certainly negligible. No other experimental effect which could explain the difference between theoretical and experimental values of the constant A has presented itself to date, so that the reason for the discrepancy remains unresolved.

5.5 Measurements at Oblique Incidence

Consider the arrangement depicted in Fig. 10, where the zero balance position is chosen so that the beam impinges on the surface at an angle of incidence θ. If the re-emitted momentum has magnitude p_r and is inclined at an angle φ to the normal, on the opposite side from the incident beam, the torque exerted on the balance is

$$C = p_i \ell + p_r \ell \sec \theta \cos \varphi \tag{5.3}$$

where ℓ is the perpendicular distance from the beam axis to the torsion fiber (i.e., the moment arm measured by the cathetometer. The effective force, therefore, is

$$p = \frac{C}{\ell} = p_i + p_r \sec \theta \cos \varphi \tag{5.4}$$

In the case of complete accommodation

$$p_i = \dot{\mu}_{FM} (1 + \varphi_2)$$

$$p_r = \frac{2}{3} \dot{\mu}_{FM} (1 + \varphi_1) \tag{5.5}$$

$$\varphi = 0$$

so that

$$p = \dot{\mu}_{FM} (1 + \frac{2}{3} \sec \theta + \varphi_2 + \frac{2}{3} \varphi_1 \sec \theta)$$

$$= \frac{1}{3} \dot{\mu}_{FM} (3 + 2 \sec \theta + 3\varphi_2 + 2\varphi_1 \sec \theta) \tag{5.6}$$

and

$$p_R = 1 + \frac{3\varphi_2 + 2\varphi_1 \sec \theta}{3 + \sec \theta} \tag{5.7}$$

By comparing Eq. (5.6) with the corresponding one for normal incidence we see that the free molecular value of p at oblique incidence is larger than that at normal incidence by a factor $(3 + 2 \sec \theta)/5$. Also, the force perturbation φ is now a different weighted mean of φ_2 and φ_1 so that by comparing measured force perturbations at normal and oblique incidence it should be possible to determine φ_2 and φ_1 separately. Unfortunately, the method is not a very sensitive one. The maximum value of θ which it is convenient to use in the present apparatus is about 40°, so that $\sec \theta \sim 1.3$. Hence even if $\varphi_2 = 2\varphi_1$ the difference between the force perturbations at normal and oblique incidence is only about 4 per cent, which is about the accuracy to which the perturbation can be measured.

Results obtained at an angle of incidence of 40.4° are shown in Fig. 11. Since the preceding analysis assumes complete accommodation only argon was used in these measurements. The increase in the free molecule value of p corresponds fairly accurately to the expected value for diffuse reflection. The curve drawn in on the figure is the best fit asymptote to

the normal incidence results at the origin and it appears to fit the oblique incidence results equally well. In other words, the difference between φ_2 and φ_1 is too small to be detected in these experiments, a conclusion which is supported by Willis and Fitzjarrald's calculations for nearly free molecule flow which yield a value of φ_2/φ_1 of about 1.15. Experimentally Scott, et.al.,[7] made measurements of the distribution functions of non-ideal molecular beams for various values of the same Knudsen number. Unfortunately, only the shape of the distribution function and not its relative total intensity was measured in these experiments. Nevertheless it is possible from their results to calculate values of $\dot{\mu}/\dot{m}$ by taking the appropriate moment of their experimental distribution curves. For argon at a source Knudsen number of 1.04 the value of $\dot{\mu}/\dot{m}(2RT)^{1/2}$ we calculated from their results is 1.41. In the case of free molecule flow the theoretical value is $(9\pi/16)^{1/2}$, or 1.33. (See Eq. 4.2.) Hence $(1 + \varphi_2)/(1 + \varphi_1) = 1.06$. From our own data at $K = 1.04$ (Fig. 8), we find $(3\varphi_2 + 2\varphi_1)/5 = 0.19$. Hence

$$\varphi_2 = 0.22$$

$$\varphi_1 = 0.16$$

and $\varphi_2/\varphi_1 = 1.46$

This ratio is considerably higher than Willis' theoretical value mainly because we have used our experimental value of φ which is lower than the value predicted by Willis' theory. Morton[3] has pointed out that his theory predicts changes in shape of the speed distribution function fairly well but that his calculated change in intensity is appreciably too large (compared, presumably, to Scott's measurements of beam intensity which, unfortunately, have not yet been published). If so, the same is probably true of Willis' theory.

5.6 The Effect of a Non-Ideal Beam on Previous Results

Using the values of φ_2 and φ_1 obtained above for a Knudsen number of 1.04 it is instructive to calculate the effect of a non-ideal beam on the value of σ'' deduced from measurements of momentum transfer at this value of K when interpreted according to the simple formula (2.10), which, it should be recalled, was derived assuming an ideal Maxwellian beam. It is shown in the Appendix that when non-ideal effects are taken into account Eq. (2.10) becomes

$$\frac{p}{p_o} - 1 = \frac{2}{5}\sigma''\left[\left(\frac{T_w}{T}\right)^{1/2} - 1\right]\left\{\frac{1+\varphi_1}{1+\frac{3\varphi_2+2\varphi_1}{5}+\frac{2}{5}(1-\sigma'')(\varphi_2-\varphi_1)}\right\} \tag{5.9}$$

For argon ($\sigma'' = 1$) the term in curly brackets has the value 0.966 at $K = 1.04$, and for helium ($\sigma'' = 0.5$) the value is 0.955. Thus the error involved in using the simple formula (2.10) is only about 1 percent if the source pressures of the two gases are adjusted to give the same source Knudsen number. If, however, the source pressures are the same for the two gases (this was usually the case in the previous work) the value of K^{-1} for helium will be only 0.381, which from our data gives $(3\varphi_2 + 2\varphi_1)/5 = 0.10$. If we assume that $\varphi_2/\varphi_1 = 1.46$ as before, we find

$$\varphi_2 = 0.114$$

$$\varphi_1 = 0.071$$

and the term in curly brackets in Eq. (5.9) has the value of 0.973, which again only differs from the value for argon by about one percent. Since one cannot hope to measure σ'' to an accuracy of 1 per cent with the present apparatus (the scatter in Stickney's data was typically ±5 percent),

it seems that one would be justified in using the simple formula (2.10) even at such low Knudsen numbers. Most of Stickney's measurements were made using a slit source so that the above calculation cannot be expected to apply directly. However, the widths of the slits used (0.003" and 0.007") were much smaller than the diameter of the circular aperture employed in the present investigation and it is therefore fair to assume that in this case non-ideal beam effects were even smaller, and the use of Eq. (2.10) even more justifiable. We conclude therefore that Stickney's measured values of σ'' do not require any correction for non-ideal beam effects.

6.0 DISCUSSION AND CONCLUSIONS

The experiments described here have demonstrated the feasibility of placing measurements of momentum transfer in gas-surface interactions on an absolute basis, so that future investigations will not be obliged to rely, as did earlier work, on purely relative measurements.

One of the foremost requirements for absolute measurements is a knowledge of the properties of the incident molecular beam and these have been studied in some detail for the particularly important case of a well collimated beam emanating from a circular source orifice. Special attention has been paid to the evaluation of the mass and momentum fluxes in such a beam over a range of source Knudsen numbers from about 30 to 0.3. This includes the whole of the "nearly free molecular" flow regime of rarefied gas dynamics for which a number of approximate theories are available. The experimental results are in closest agreement with the calculations of Willis and Fitzjarrald which take into account the variation of the molecular distribution function over the area of the source aperture but the discrepancy between the measured and calculated perturbations of mass and momentum flux from their free molecule values is still about 80 percent.

This could probably be reduced by correcting the calculations to the appropriate Prandtl number of 2/3 rather than unity; however, there still appears to be a definite disagreement between theory and experiment for which there is no immediately obvious explanation. One possibility is that the discrepancy is in some way associated with the use in the calculations of the Krook model equation rather than the true Boltzmann equation, but this has by no means been established.

The diffuse reflection of helium and argon atoms from a platinum surface under the conditions of the present experiment has been demonstrated in two ways:

1) by making absolute measurements of normal momentum transfer to the surface from a molecular beam operated at sufficiently high source Knudsen numbers for intermolecular collisions to be neglected (helium) or by extrapolating experimental results to such conditions (argon);

2) by comparing the momentum transfer to a platinum surface over a wide range of source conditions with that to a momentum trap specifically designed to give completely diffuse reflection.

In each case the measured momentum transfer to the platinum surface agreed with the value for diffuse reflection to within about ±1.5 percent.

Although this result is not particularly surprising, its accurate verification is valuable; in particular, it justifies one of the most important assumptions made in interpreting the earlier measurements of normal momentum transfer to a heated surface. Although the present measurements were restricted to the case of a platinum surface, the fact that Stickney's results for other surfaces such as tungsten and aluminum were very similar to those for platinum and gave almost identical values of σ'' suggests that surface contamination plays a dominant role in all these measurements and that the physical nature of the test surface is unimportant. In experiments

performed under much cleaner vacuum conditions this may of course no longer be true. Datz and Moore[8] have observed significantly specular reflection of modulated beams of helium and deuterium from clean platinum surfaces. If the necessary improvement in cleanliness can be achieved in the present apparatus it would be interesting to see if such an effect could be detected by momentum transfer measurements.

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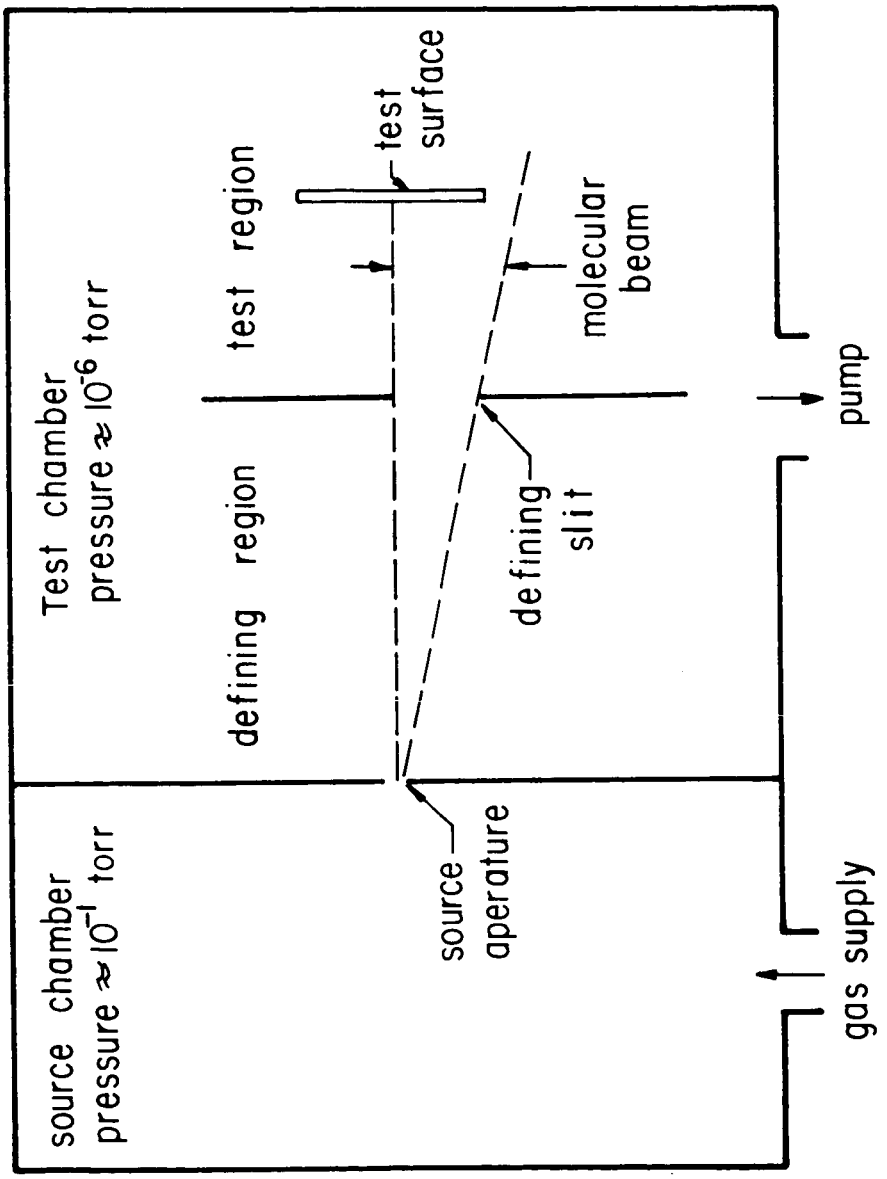

FIG. 1 SCHEMATIC DIAGRAM OF MOLECULAR BEAM APPARATUS

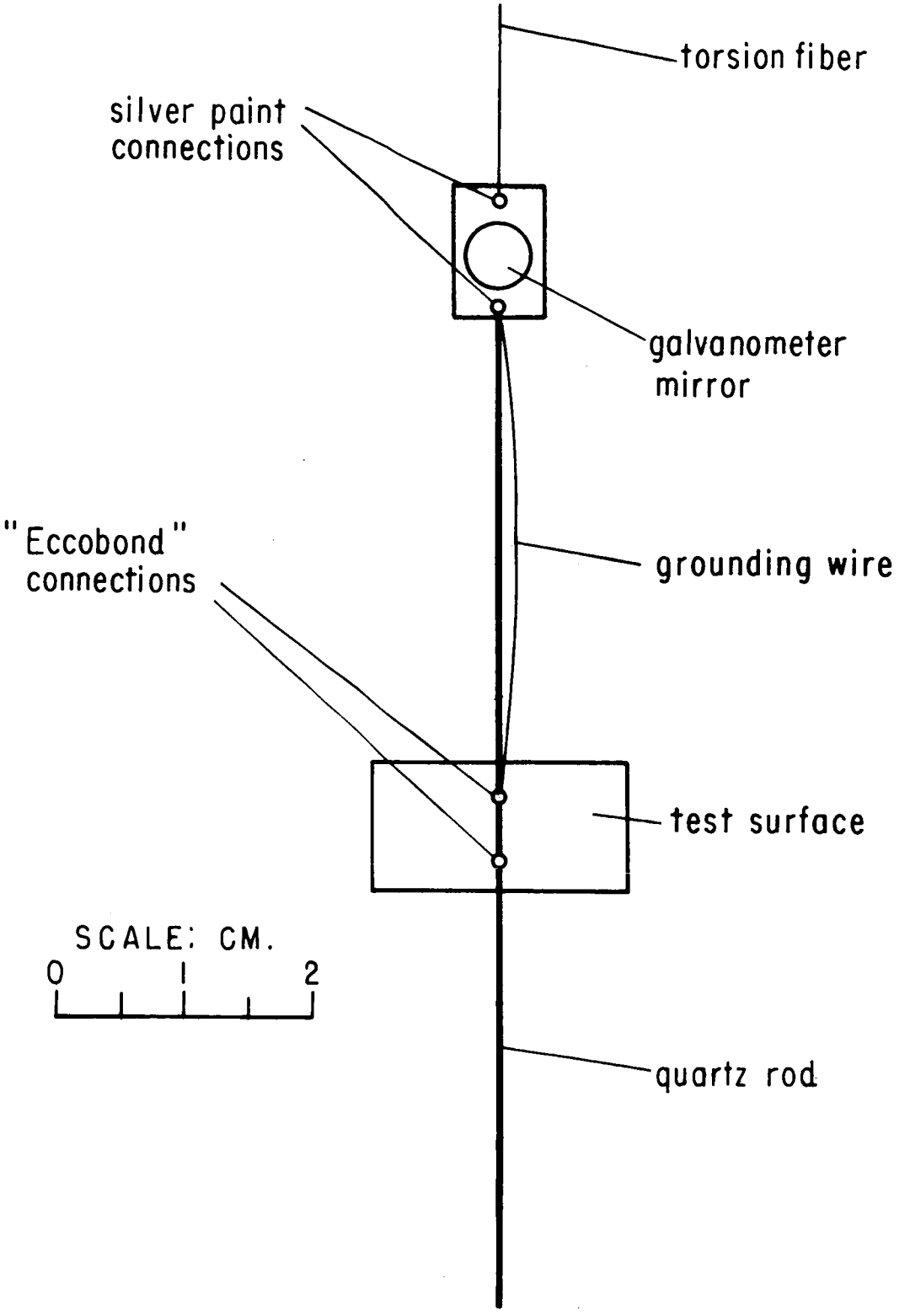

FIG. 2 CONSTRUCTION OF TORSION BALANCE

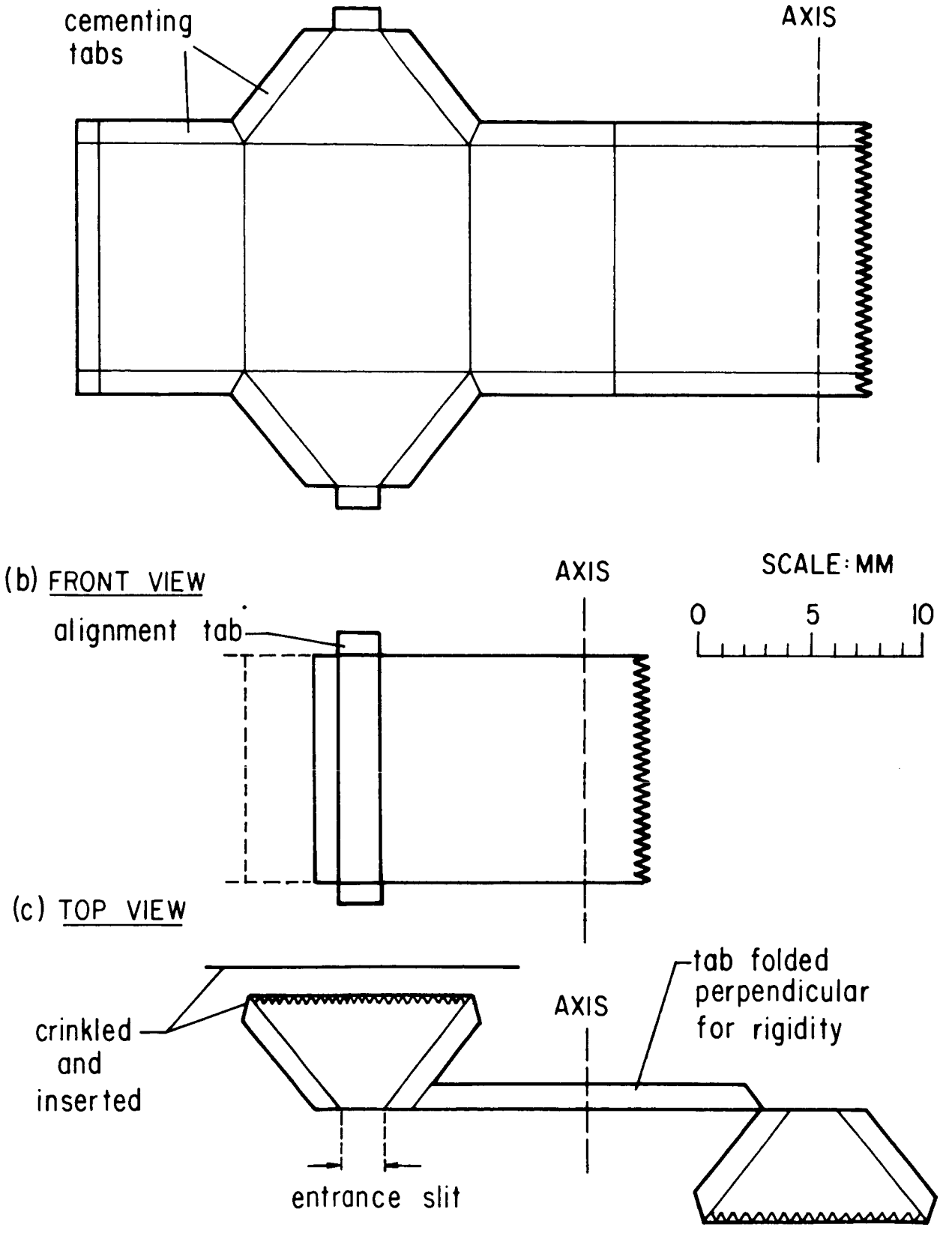

FIG. 3 CONSTRUCTION OF MOMENTUM TRAP

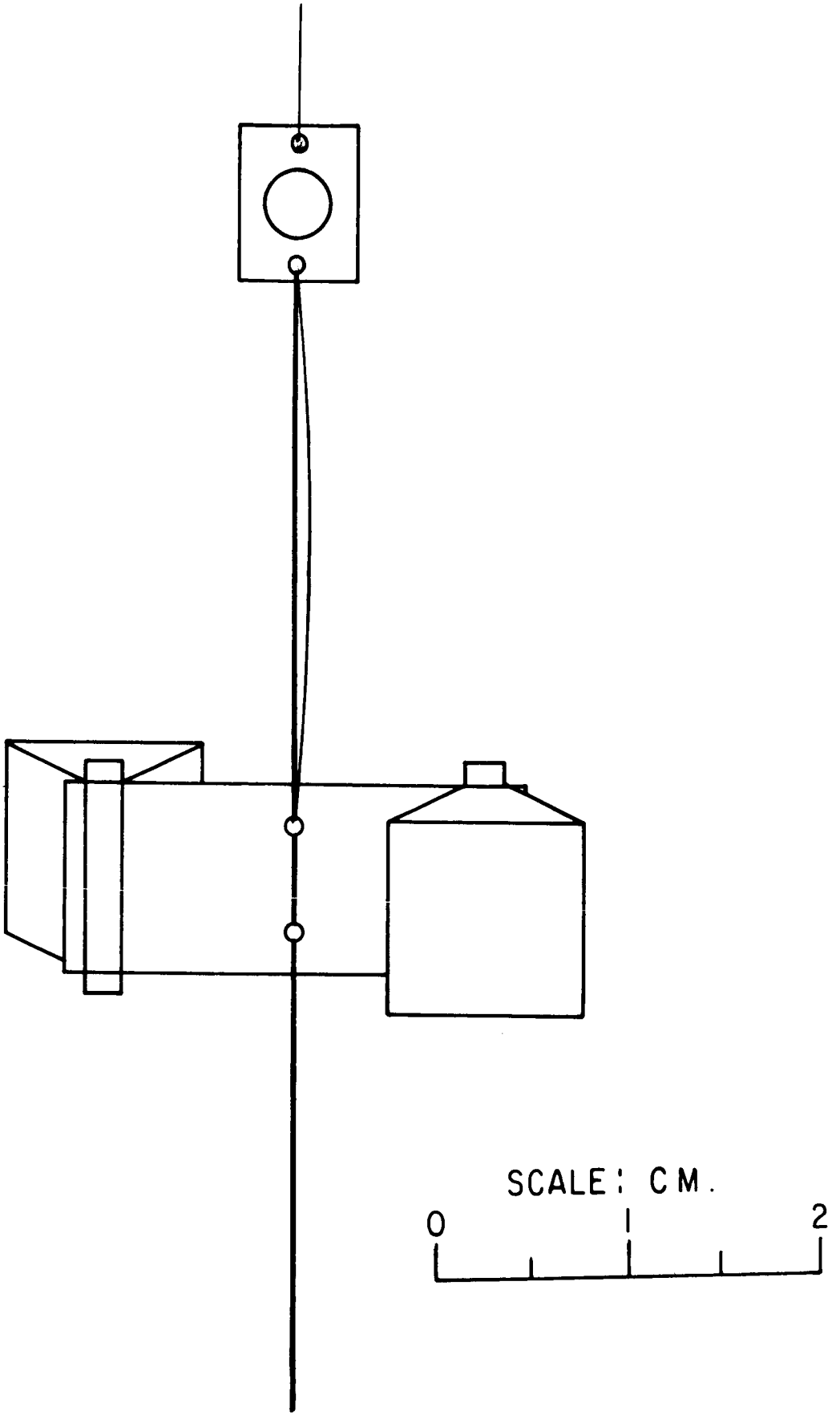

FIG. 4 MOMENTUM TRAP BALANCE

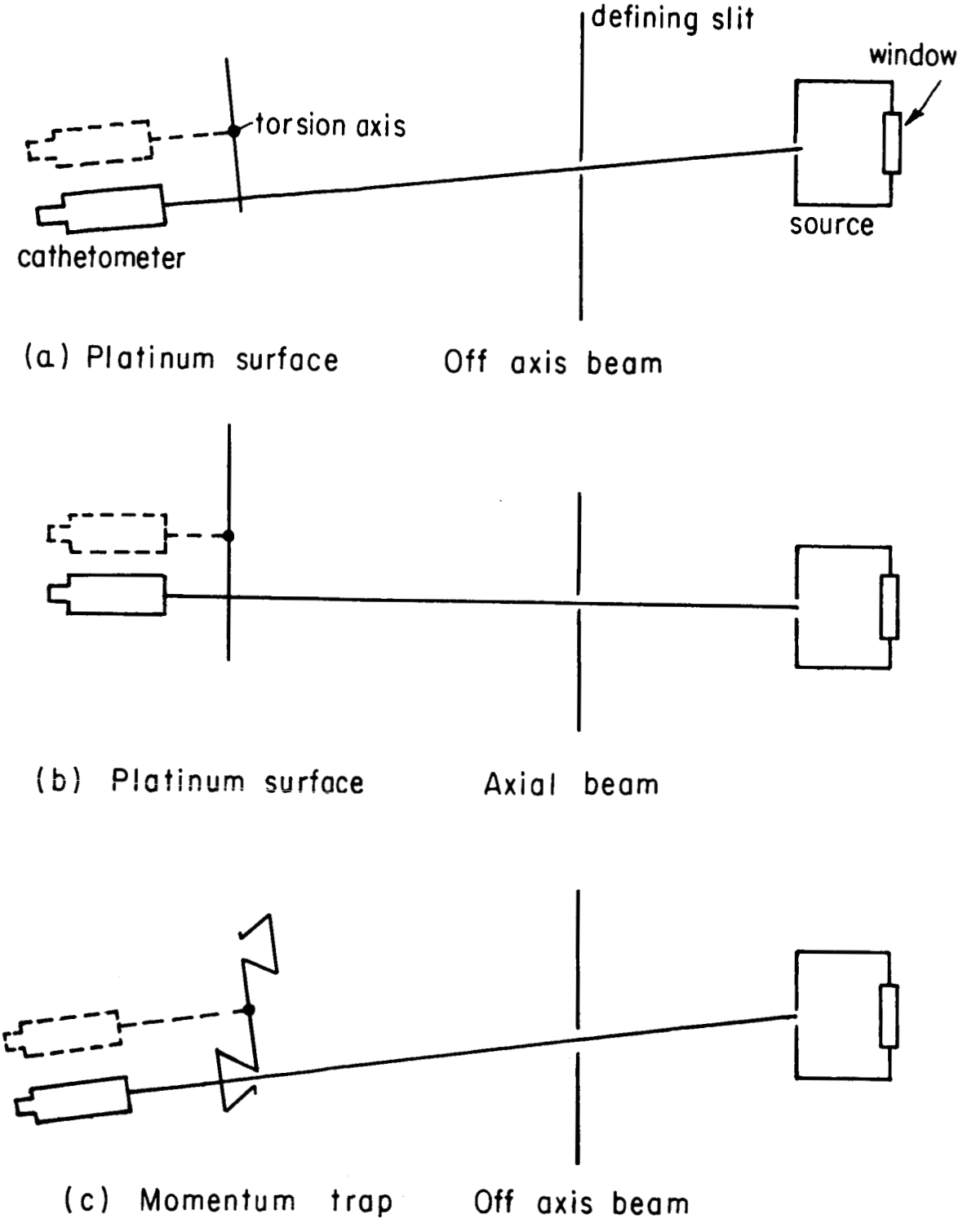

FIG. 5 ALIGNMENT CONFIGURATIONS

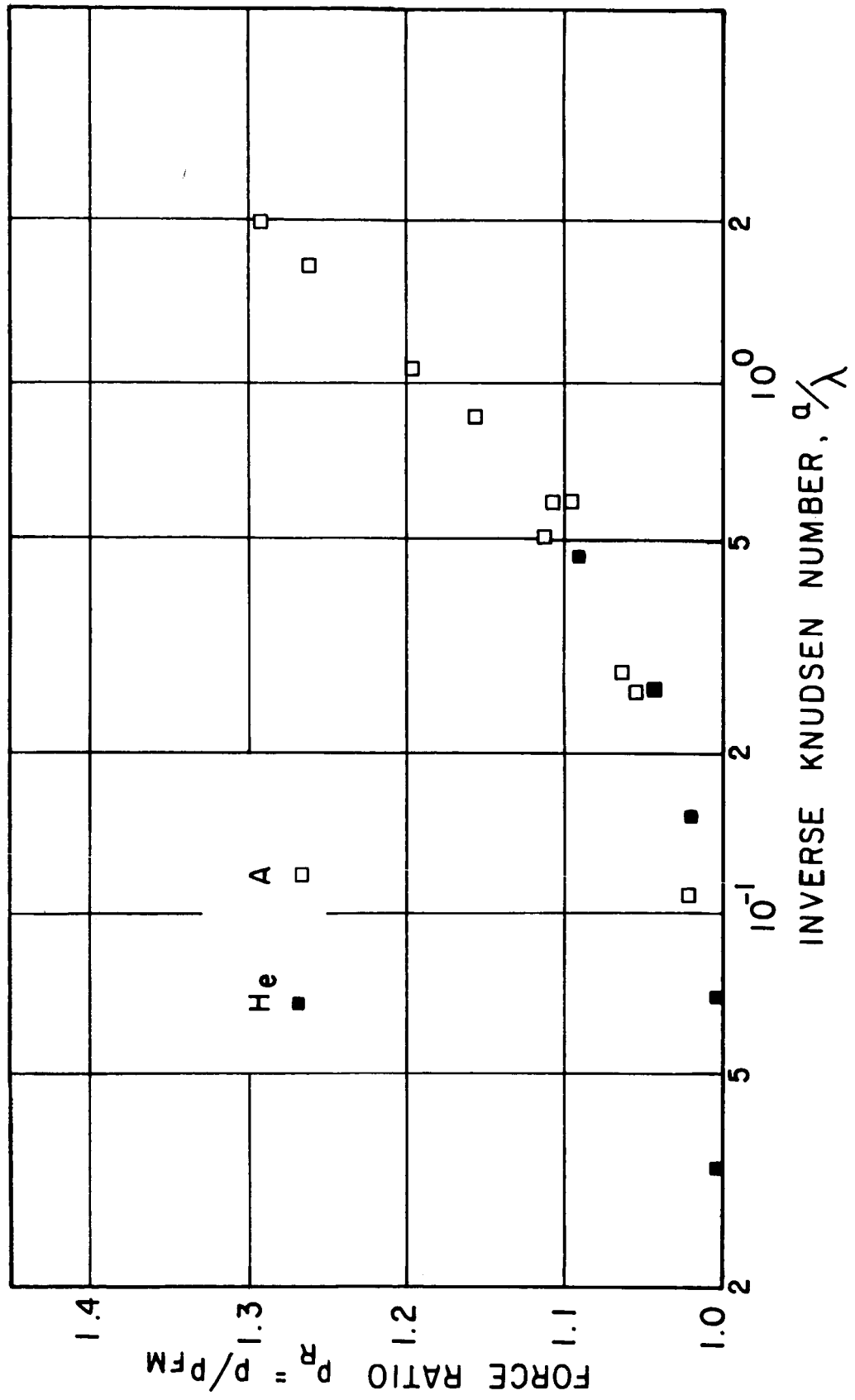

FIG. 6 EXPERIMENTAL RESULTS FOR MOMENTUM TRAP

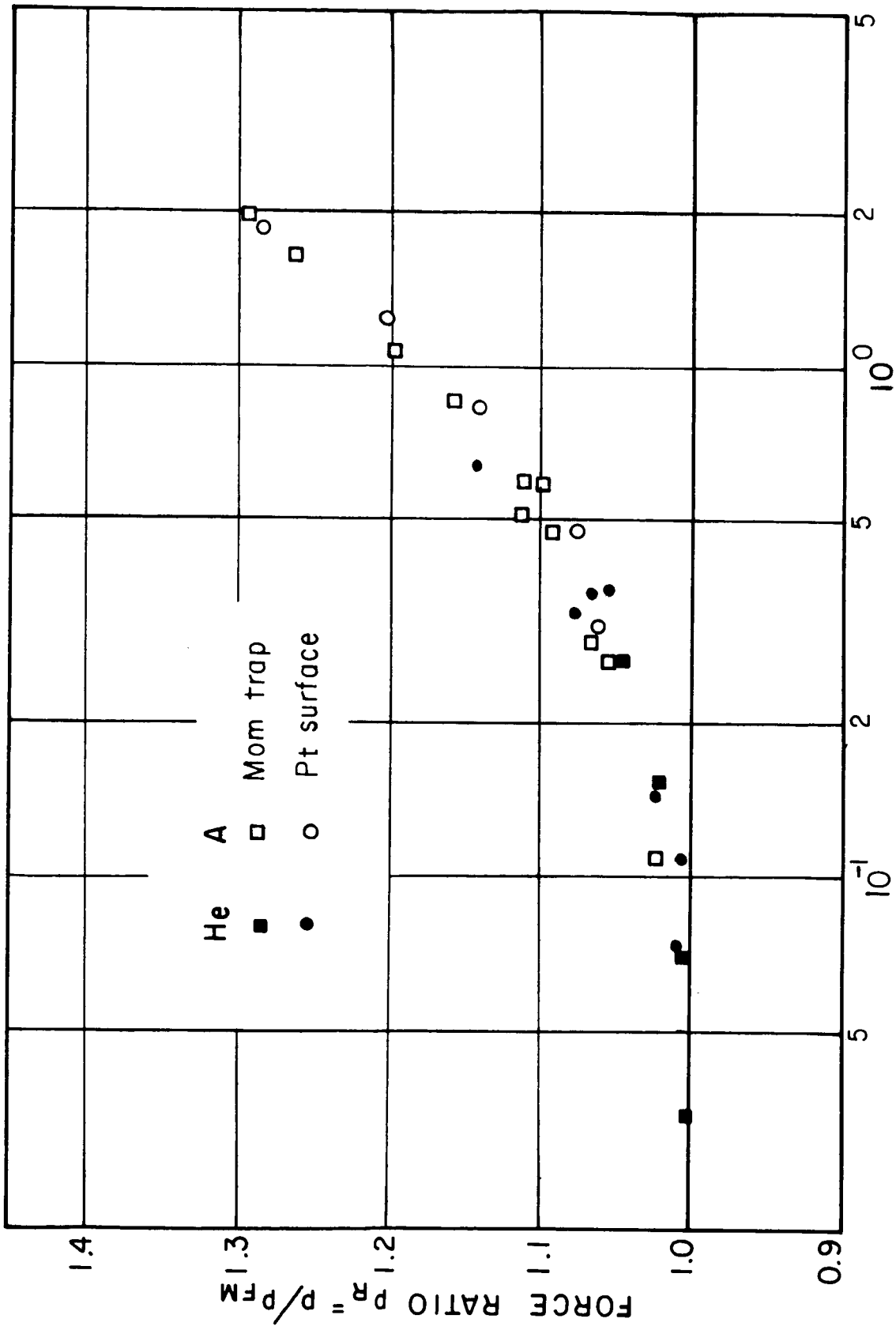

FIG.7 EXPERIMENTAL RESULTS FOR MOMENTUM TRAP AND PLATINUM SURFACE

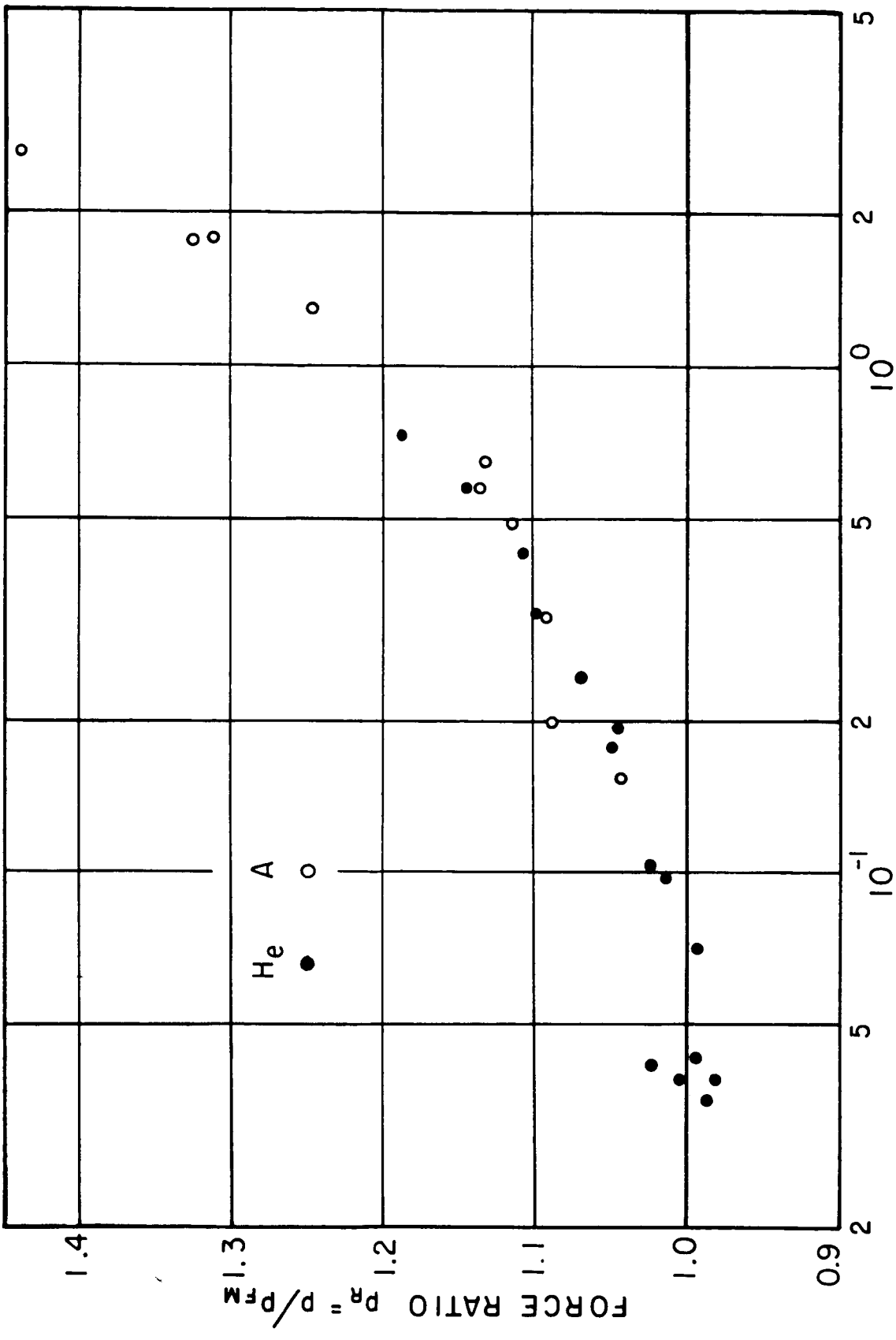

FIG. 8a MOMENTUM TRANSFER TO PLATINUM SURFACE USING AXIAL BEAM

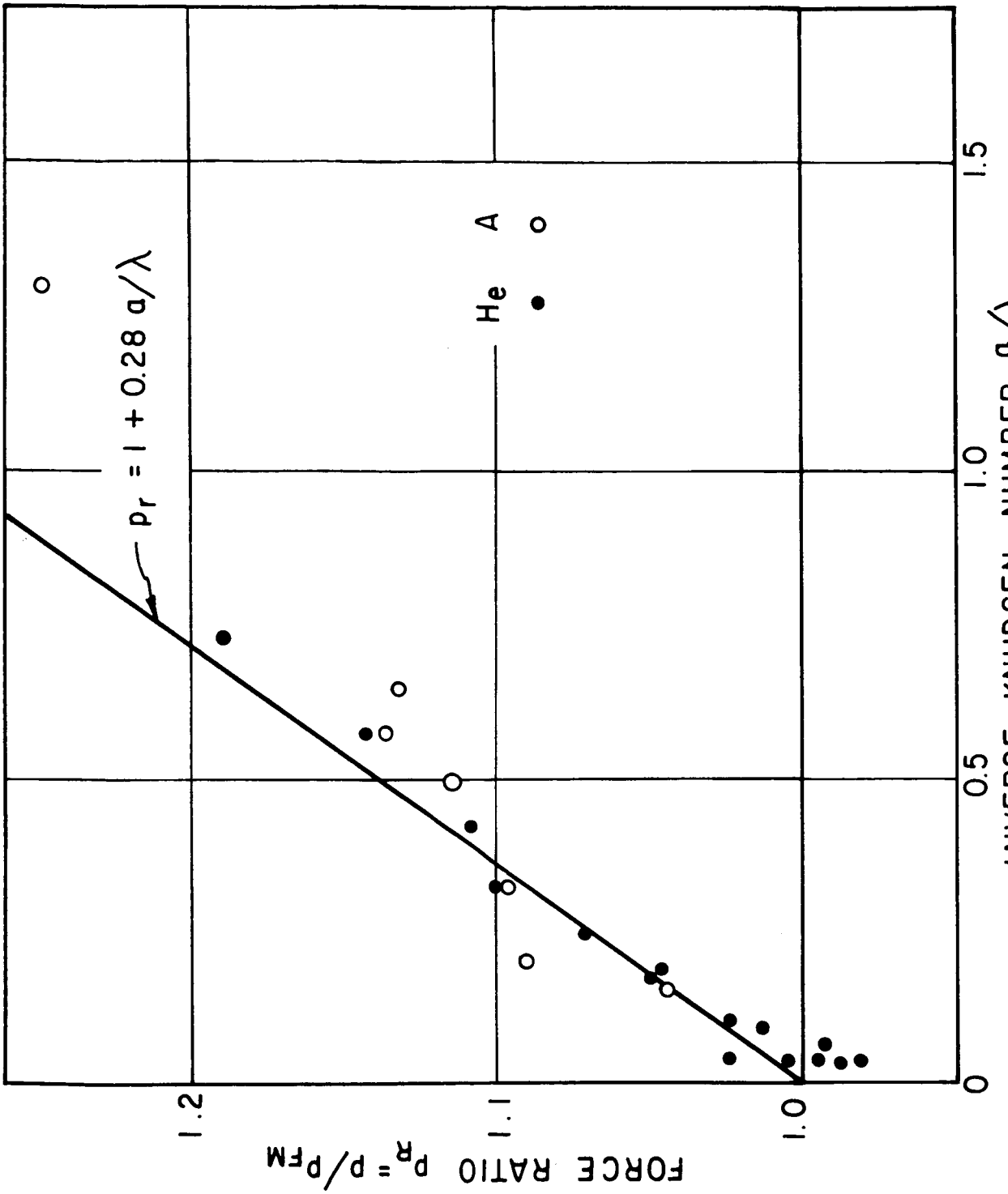

FIG. 8b MOMENTUM TRANSFER TO A PLATINUM SURFACE USING AXIAL BEAM

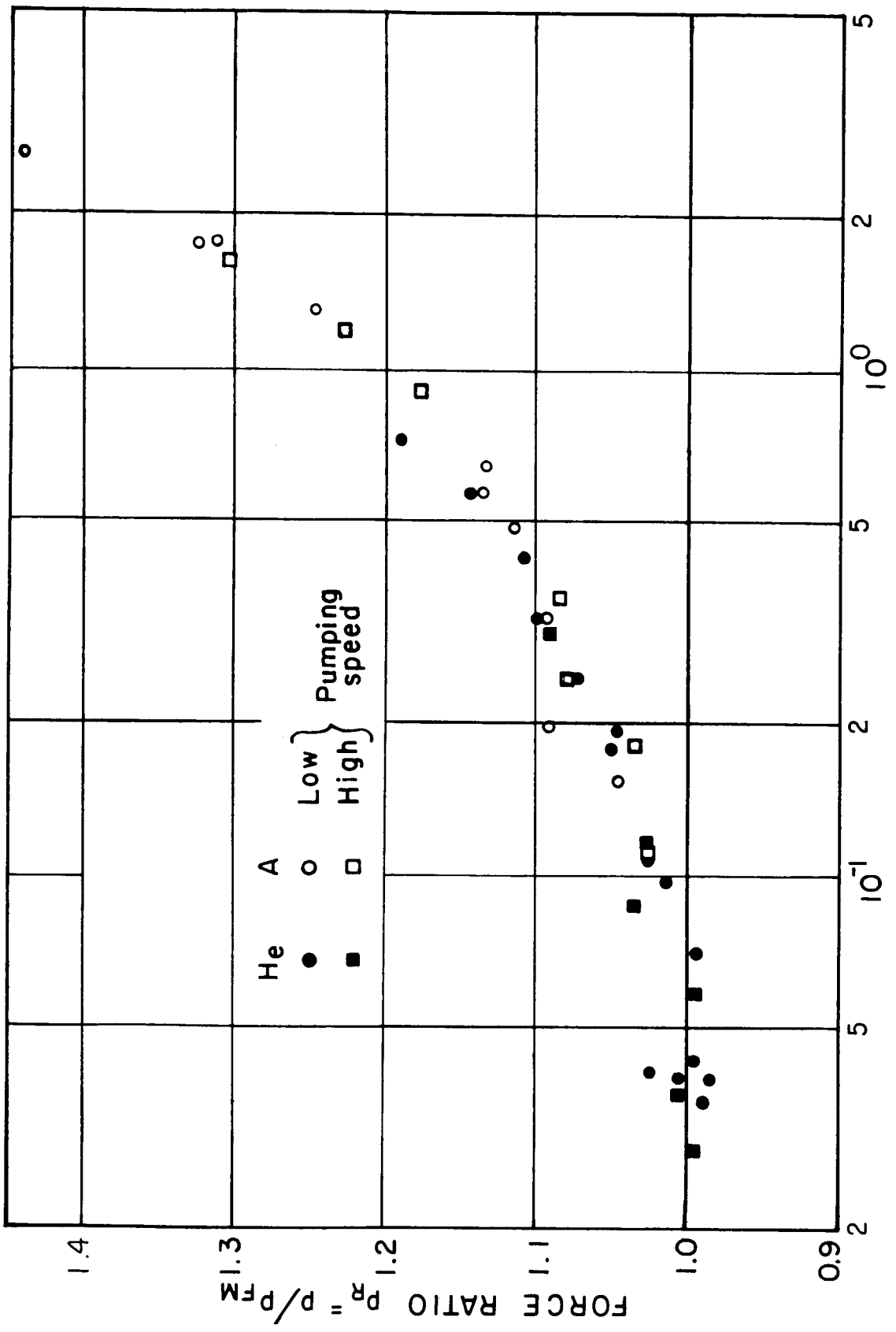

FIG.9 MOMENTUM TRANSFER TO PLATINUM SURFACE AT TWO DIFFERENT PUMPING SPEEDS

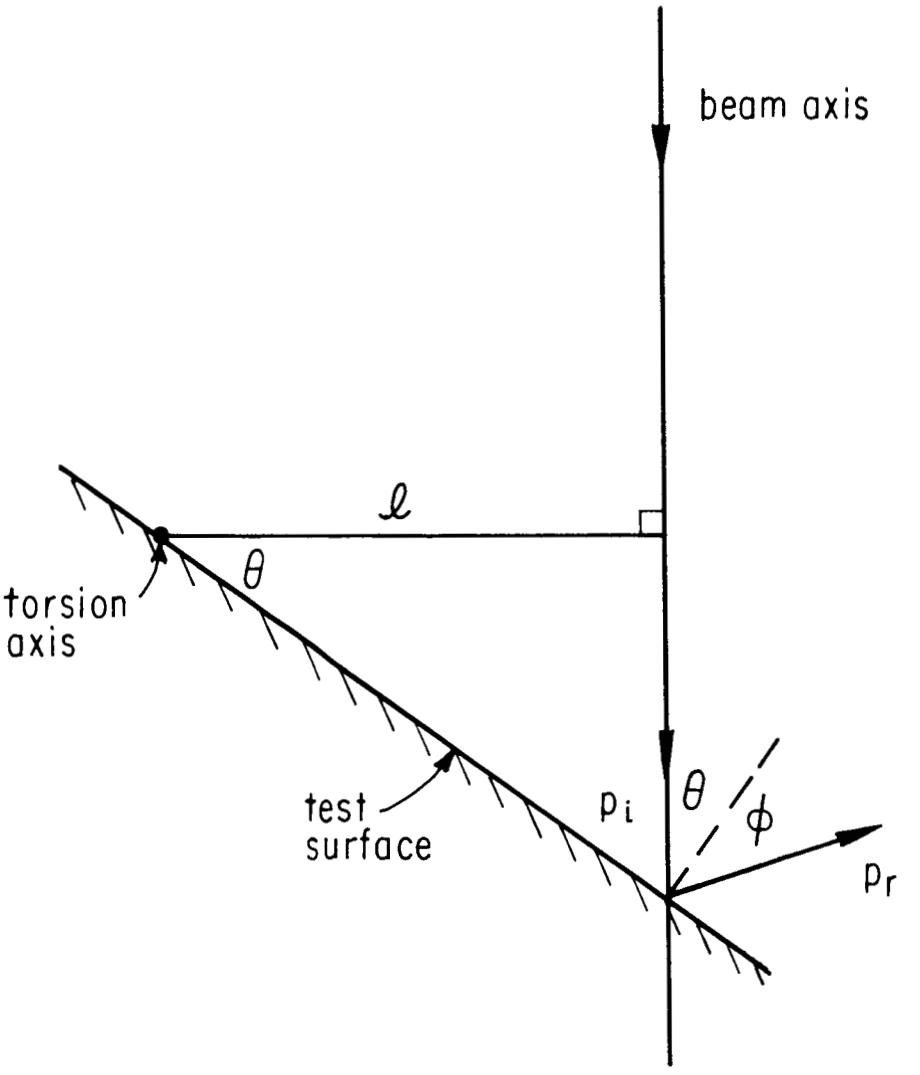

FIG. 10 EXPERIMENTAL ARRANGEMENT FOR OBLIQUE INCIDENCE MEASUREMENTS

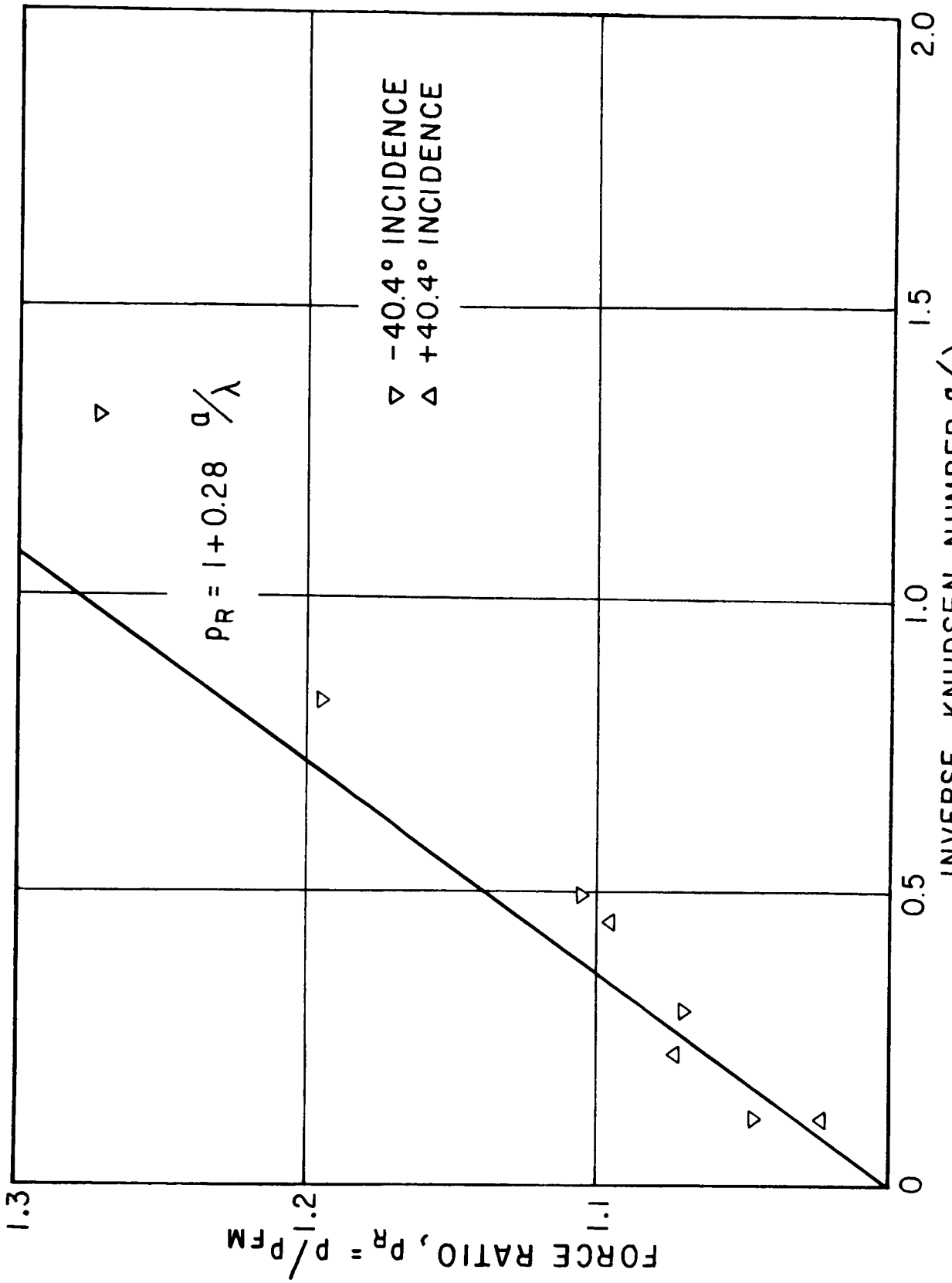

FIG.11 MOMENTUM TRANSFER TO A PLATINUM SURFACE AT OBLIQUE INCIDENCE. ARGON DATA

APPENDIX

Consider a well collimated molecular beam from a source at temperature T impinging at normal incidence on a diffusely reflecting surface at temperature T_w. We shall work in terms of the model interaction described on p. 16 in which a fraction σ'' of the incident molecules are assumed to be completely accommodated to the temperature T_w while the rest are reflected in multi-specular fashion with no energy accommodation.

The incident momentum and mass fluxes are

$$\dot{\mu} = \dot{\mu}_{FM} (1 + \varphi_2) = p_i \qquad \text{(A-1)}$$

$$\dot{m} = \dot{m}_{FM} (1 + \varphi_1) \qquad \text{(A-2)}$$

The normal momentum flux carried away by molecules of the first category is (from Eq. 4.4)

$$p_r^{(1)} = \sigma'' \left(\frac{\pi R T_w}{2} \right)^{1/2} \dot{m}$$

$$= \sigma'' \left(\frac{\pi R T_w}{2} \right)^{1/2} \dot{m}_{FM} (1 + \varphi_1) \qquad \text{(from A-2)}$$

$$= \sigma'' \left(\frac{\pi R T_w}{2} \right)^{1/2} (1 + \varphi_1) \times \frac{2}{3} \left(\frac{2}{\pi R T} \right)^{1/2} \dot{\mu}_{FM} \qquad \text{(from 4.2)}$$

$$= \frac{2}{3} \sigma'' \dot{\mu}_{FM} (1 + \varphi_1) \left(\frac{T_w}{T} \right)^{1/2} \qquad \text{(A-3)}$$

That carried away by molecules of the second kind is from (4.7)

$$p_r^{(2)} = (1 - \sigma'') \times \frac{2}{3} \dot{\mu} = \frac{2}{3} (1 - \sigma'') \dot{\mu}_{FM} (1 + \varphi_2) \qquad \text{(A-4)}$$

The total force on the surface is therefore

$$p = p_i + p_r^{(1)} + p_r^{(2)}$$

$$= \dot{\mu}_{FM} \left[(1+\varphi_2) + \frac{2}{3}\sigma'' (1+\varphi_1) \left(\frac{T_w}{T} \right)^{1/2} + \frac{2}{3}(1-\sigma'')(1+\varphi_2) \right] \quad (A\text{-}5)$$

In the special case where $T_w = T$ denoted by subscript o

$$p_o = \dot{\mu}_{FM} \left[(1+\varphi_2) + \frac{2}{3}\sigma'' (1+\varphi_1) + \frac{2}{3}(1-\sigma'')(1+\varphi_2) \right] \quad (A\text{-}6)$$

which may easily be verified to be yet another way of writing Eqs. (4.8) and (4.9). Hence

$$p-p_o = \frac{2}{3}\sigma'' (1+\varphi_1) \left[\left(\frac{T_w}{T} \right)^{1/2} - 1 \right] \dot{\mu}_{FM} \quad (A\text{-}7)$$

Dividing by the expression for p_o given in Eq. (4.9), we finally obtain

$$\frac{p-p_o}{p_o} = \frac{2}{3}\sigma'' \left[\left(\frac{T_w}{T} \right)^{1/2} - 1 \right] \left\{ \frac{1+\varphi_1}{1 + \frac{3\varphi_2+2\varphi_1}{5} + \frac{2}{5}(1-\sigma'')(\varphi_2-\varphi_1)} \right\} \quad (A\text{-}8)$$

which is the required result.